REMARKS

Reconsideration and allowance of this application are respectfully requested in light of the foregoing amendments and the following remarks.

Claim Status

Claim 1 is amended. Claims 1-12 and 14-18 are pending. No new matter was added.

§102/103 Claim Rejections

Claims 1-9, 12 and 14-18 stand rejected under 35 U.S.C. §102(b) as being anticipated by U.S. Patent No. 5,840,190 (hereinafter Scholander). Additionally, Claims 10 and 11 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Scholander. Applicant traverses.

To anticipate a claim under 35 U.S.C. §102(b), a single source must contain all of the elements of the claim. *See Hybritech Inc. v. Monoclonal Antibodies, Inc.,* 802 F.2d 1367, 1379, 231 USPQ 81, 90 (Fed. Cir. 1986); *Atlas Powder Co. v. E.I. du Pont De Nemours & Co.,* 750 F.2d 1569, 1574, 224 USPQ 409, 411 (Fed. Cir. 1984); *In re Marshall,* 578 F.2d 301, 304, 198 USPQ 344, 346 (C.C.P.A. 1978). Missing elements may not be supplied by the knowledge of one skilled in the art or the disclosure of another reference. *See Structural Rubber Prods. Co. v. Park Rubber Co.,* 749 F.2d 707, 716, 223 USPQ 1264, 1271 (Fed. Cir. 1984). Where a reference discloses less than all of the claimed elements, an Examiner may only rely on 35 U.S.C. §103. *See*

Titanium Metals Corp. v. Banner, 778 F.2d 775, 780, 227 USPQ 773, 777 (Fed. Cir. 1985).

Contrary to the Examiner's belief, Scholander fails to anticipate the instant invention. The Examiner maintains his rejection of independent claims 1 and 12 despite the fact that Scholander does not qualify as prior art to the present invention. Applicant has amended the language of claim 1 in order to clarify the meaning of that claim. Looking to the Examiner's comments on page 4, paragraph 15 of the current office action, the Examiner states that he "considers the surface modified membrane of Scholander to be an asymmetric membrane." With all respect due to the Examiner, Applicant simply cannot understand how, in light of the definition given by the Examiner himself in paragraph 15, which relates to morphological and structural aspects, one having skill in the art could consider a membrane having one of the surfaces modified to be an asymmetric membrane.

Applicant would first like to point out what, in the context of the instant invention, must be understood to be an asymmetric membrane. Applicant would like to draw the Examiner's attention to page 1, paragraph 2 of the current specification where it clearly describes that an integrally asymmetric membrane is understood to be one having a separating layer and a supporting layer, wherein both layers are bound together in an integral unit. This is the meaning of the term "asymmetric membrane" as used in the context of the instant invention, and the disclosure in the prior art must be assessed in light of this statement. The current patent application uses the term "asymmetric

membrane" purely in the sense of an asymmetry with respect to the structural and morphological conditions within the membrane wall, i.e. with respect to pore structure, pore size, etc.

Furthermore, the way the term "asymmetric membrane" is used in the context of the present invention is in agreement with the use of the term in literature on membrane technology. Applicant refers to the excerpt of the textbook of M.C. Porter: "Handbook of Industrial Membrane Technology" (which was submitted to the Examiner along with Applicant's response to the previous office action on May 24, 2010) and specifically to the paragraph "Asymmetric Microporous Membranes" on page 12. Applicant also encloses the article, "Terminology for Membranes and Membrane Processes" (enclosure 1), in which recommendations for the terminology for membranes are listed. According to this article, an asymmetric membrane is a membrane constituted of two or more <u>structural</u> planes of non-identical morphology, i.e. asymmetric relates to morphological or structural aspects of the membrane only.

Applicant also refers to the definitions given in various textbooks such as:

a. L.J. Zeman: "Microfiltration and Ultrafiltration" (enclosure 2). Looking to page 108 where it states that ultrafiltration and microfiltration membranes often have a cross-sectional pore size gradient such that their <u>structure</u> displays a character of gradient anisotropy (which is equivalent to asymmetry).

b.   R.E. Kesting: "Synthetic Polymeric Membranes" (enclosure 3). Looking to page 265 it is explained in Chapter 7.6 that "…asymmetry refers to a <u>structure</u> in which a thin dense skin layer is integrally bonded in series with a thick porous substructure", i.e. again reference is made to purely structural aspects.

c.   M. Mulder: "Basic Principles of Membrane Technology" (enclosure 4). Looking to the last paragraph on page 12, one reads that asymmetric membranes consist of a dense top-layer and a porous sub-layer.

In contrast to the asymmetric membranes as defined in the instant application and as defined in the literature on membranes illustrated above, the membranes of Scholander are "only" surface modified. However, the surface modification occurring on the membranes of Scholander does not relate at all to a modification of the surface with respect to structural or morphological conditions. The surface modification of the membranes of Scholander only has the effect to give functional groups onto the membrane surface to which antithrombotic compounds are immobilized/covalently bound (see Scholander, e.g. claim 1). The motivation of Scholander is to improve the biocompatibility of the membrane surface (see Column 3, Line 65 to Column 4, Line 9), i.e, the compatibility of the surface with respect to the organism's defence system such as the activation of the coagulation, complement and immune systems. (Column 1, Lines 36-40). For improving biocompatibility, membrane surfaces are modified with respect to their negative or positive charge (Column 1, Lines 50-55) or functional groups are generated to which ionically or covalently bonded biologically active compounds can

be coupled (Column 2, Lines 18-23). Thus, the endeavour of these methods is to alter the membrane surface with respect to their physico-chemical properties, but not to alter the membrane structure or morphology to make one side different from the other.

As is disclosed by Scholander, several prior art modification methods pose a serious disadvantage in that they may alter the physical properties of a membrane, even such that even the pores are blocked. (Column 2, Lines 52-58).

Scholander wants to avoid these draw-backs (Column 2, Lines 59-61). This means that the membranes of Scholander do not show an alteration of the physical properties as a result of the surface modification which also means that the membrane structure is not affected by the surface modification. The surface modification of Scholander only relates to the functionality of the membrane's surface with respect to biocompatibility. This, however, as explained above, has absolutely nothing to do with asymmetry and more specifically is in stark contrast to asymmetry as used in the context of the instant invention.

As a result, Scholander does not relate to asymmetric membranes, let alone to asymmetric membranes having a separating layer wherein a polyelectrolyte with negative fixed charges, but having no positive fixed charges, is bound in this separating layer.

The Examiner further states in paragraph 15 of the current office action that the process steps of the invention are disclosed in the prior art. In paragraph 14, the Examiner states that the applicant has argued that Scholander does not disclose a step in which the first and/or second surface of a shaped object is brought into contact with a precipitant system. However, in this respect the Examiner unduly shortens our argument. In Applicant's response to the previous office action, Applicant did not allege that Scholander does not disclose a step in which the first and/or second surface of a shaped object is brought into contact with a precipitant system. Instead, Applicant stated that Scholander does not disclose a step in which the first and/or second surface of a shaped object is brought into contact with a precipitant system which is such that a membrane results having a separating layer on the first and/or second surface. Thus, according to the process of the instant invention, the precipitant system must meet the condition that it must be such that a separating layer indeed is formed. Moreover, the precipitant system which is causative for the separating layer must contain a polyelectrolyte with negative fixed charges, but no positive fixed charges.

Only in cases where there exists a separating layer covering a supporting layer will an asymmetric membrane result (see definitions above). Moreover, only in cases where the precipitant system which causes the separating layer to be formed is comprised of a polyelectrolyte with negative fixed charges, but no positive fixed charges, as is required by the present invention, will an improvement in separation efficiency be realized.

As stated in our previous response, there is no implicit disclosure within Scholander that a membrane is formed having a separating layer on the first and/or second surface, as is alleged by the Examiner. Applicant must assume that because Scholander describes a process in which a membrane is precipitated from a casting solution in a non-solvent (column 4, line 38 – column 5, line 10) the Examiner draws the conclusion (incorrectly) that an asymmetric membrane with a separating layer and a supporting layer is produced. The Examiner provides no further explanation.

Unfortunately for the Examiner, one skilled in the art would clearly know that this conclusion is not appropriate. Just because a precipitation is effected, one cannot conclude that inevitably an integrally asymmetric membrane with a separation layer is obtained. One skilled in the art knows that numerous factors during membrane production contribute to the end product achieved. Thus, depending on e.g. the polymer, the polymer concentration in the casting solution, the type of solvent system, the temperature of the casting solution, the precipitant system, the ratio of solvent to non-solvent in the precipitant system and the temperature of the precipitant system, a symmetrical (homogeneous) membrane, a symmetric membrane having separation layers with equal separation characteristics on both of their surfaces, asymmetric membranes having a separating layer on one or both sides or asymmetric membranes without separating layers can be obtained. From the disclosure of Scholander, one cannot say with certainty which type of membrane is produced.

In reference to claims 2-11 and 14-18, "[I]f an independent claim is not anticipated by prior art, then its dependent claims, which necessarily include the limitations of the independent claim, are not anticipated either. *Kovin Assoc. v. Extech/Exterior Technologies*, 2006 U.S. Dist. LEXIS 63250 (N.D. Ill. 2006), citing *Trintec Indus., Inc. v. Top-U.S.A. Corp.*, 295 F.3d 1292, 1296 (Fed. Cir. 2002). Thus, claims 2-11 and 14-18 are not anticipated by Scholander and should be allowed.

Regarding the rejection of claims 10 and 11 under §103, the Examiner claims to have established a prima facie case of obviousness against the instant application. MPEP § 2143 "Basic Requirements of a *Prima Facie* Case of Obviousness" states:

> To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine references teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all claim limitations.

> Regarding the third criterion, the court has stated that "to establish *prima facie* obviousness of a claimed invention, all the claim limitations must be taught or suggested by the prior art." *In re Royka*, 490 F.2d 981, 180 USPQ 580 (CCPA 1974). Applicant contends that the prior art reference Scholander fails to teach, suggest, or provides a motivation for an integrally asymmetric membrane as described in the claims of the instant invention.

The instant application is directed to a membrane having improved separation characteristics and to a method for production of such a membrane. Starting from a

membrane having an integrally asymmetric structure across its wall thickness and having at least one separating layer and a supporting layer adjoining the separating layer, the improvement is achieved for the membranes of the invention by physically binding a polyelectrolyte with negative fixed charges, but without positive fixed charges, to the separating layer, while leaving the supporting layer free from polyelectrolyte.

Such a membrane can be produced by a method as disclosed in claim 1 of the present invention. The method describes a process for creating an integrally asymmetric membrane with at least one separating layer and a supporting layer adjoining the separating layer, in which process a spinning solution is converted into a shaped object with a first and second surface (i.e. for example a hollow fiber or film) and thereafter the shaped object is brought with its first or second surface into contact with a precipitation system, wherein the precipitant system is such that formation of a membrane results in having a separating layer on said surface. This means that the precipitation system must be such and must act on the shaped object such that a separating layer is formed on the surface of the shaped object that had been brought into contact with the precipitation system.

Scholander does not disclose, either explicitly or implicitly, membranes having a separating layer. Additionally, Scholander does not disclose, either explicitly or implicitly, a method for the production of an integrally asymmetric membrane with a separating layer and a supporting layer. Moreover, Scholander fails to address features such as the separating characteristic of a membrane, let along how to improve that

separating characteristic. Instead, Scholander aims at improving the biocompatibility of the membranes (see title or column 3, line 65 to column 4, line 4).

The prior art reference or combination of references relied upon by the Examiner must teach or suggest all of the limitations of the claims. See *In re Zurko*, 111 F.3d 887, 888-89, 42 U.S.P.Q.2d 1467, 1478 (Fed. Cir. 1997); *In re Wilson*, 424 F.2d 1382, 1385, 165 U.S.P.Q. 494, 496 (C.C.P.A. 1970) ("All words in a claim must be considered in judging the patentability of that claim against the prior art."). The teachings or suggestions, as well as the expectation of success, must come from the prior art, not applicant's disclosure. See *In re Vaeck*, 947 F.2d 488, 493, 20 U.S.P.Q.2d 1438, 1442 (Fed. Cir. 1991). In this instance, from the information detailed above, it is clear that Scholander fails to teach or suggest all the limitations of Applicant's claims. Accordingly, Scholander does not disclose all of the elements of claims 10 and 11 and therefore, this rejection must fail. Thus, claims 10 and 11 are not obvious over Scholander and should be allowed.

Conclusion

In view of the foregoing, Applicant respectfully requests an early Notice of
Allowance in this application.

Respectfully submitted,

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INTERNATIONAL UNION OF PURE
AND APPLIED CHEMISTRY

MACROMOLECULAR DIVISION
COMMISSION ON FUNCTIONAL POLYMERS*
Working Party on Membrane Nomenclature†

TERMINOLOGY FOR MEMBRANES AND
MEMBRANE PROCESSES

(IUPAC Recommendations 1996)

Prepared for publication by

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Terminology for membranes and membrane processes (IUPAC Recommendations 1996)

SYNOPSIS

This project has involved assembling a basic set of terms applicable to non-living membranes and membrane processes. The terms include the key vocabulary used in the literature concerned with scientific, technical and commercial aspects of the membrane field. Processes and membranes based on synthetic or modified natural polymers as well as ceramic and metallic membranes are covered. The resultant terms and their definitions represent a core set that will allow uniform understanding and eliminate most ambiguity or confusion caused by conflicting terminology now in use. The terms and definitions selected for inclusion reflect the majority preference of the membrane community. To avoid irreconcilable differences, definitions and descriptions involving interpretations of phenomena were minimized. Some terms were deleted in cases where majority preferences were not apparent. This strategy was adopted with the understanding that the current core group of terms, which enjoy majority support, can be augmented as evolution of the field occurs.

Others contributing to this report: Professor Philippe Aptel (France), Dr. John Armor (U.S.A.), Professor Remy Audinos (France), Dr. Richard W. Baker (U.S.A.), Dr. Robert Bakish (U.S.A.), Professor Georges Belfort (U.S.A.), Dr. B. Bikson (U.S.A.), Dr. Robert G. Brown (U.S.A.), Professor Mikhailo Bryk (Ukraine), Dr. James J. Burke (U.S.A.), Professor Israel Cabasso (U.S.A.), Dr. Rey T. Chern (U.S.A.), Professor Munir Cheryan (U.S.A.), Professor E. L. Cussler (U.S.A.), Professor Robert H. Davis (U.S.A.), Dr. Thomas A. Davis (U.S.A.), Professor Enrico Drioli (Italy), Dr. D. J. Edlund (U.S.A.), Dr. Peter Eriksson (U.S.A.), Dr. Louis Erredo (U.S.A.), Dr. William Eykamp (U.S.A.), Dr. Douglas Fain (U.S.A.), Dr. R. W. Field (United Kingdom), Dr. Greg K. Fleming (U.S.A.), Professor Shintaro Furusaki (Japan), Professor George R. Gavalas (U.S.A.), Professor Bengt Hallstrom (Sweden), Professor H. B. Hopfenberg (U.S.A.), Dr. H. Philip Hsieh (U.S.A.), Professor Robert Y. M. Huang (Canada), Dr. E. Jacobs (South Africa), Dr. Kenji Kamide (Japan), Dr. Robert Kesting (U.S.A.), Professor Shoji Kimura (Japan), Professor Elias Klein (U.S.A.), Dr. Masaru Kurihara (Japan), Dr. Eric K. Lee (U.S.A.), Professor Kew-Ho Lee (Korea), Mr. Stephen A. Leeper (U.S.A.), Professor Jerry Lin (U.S.A.), Professor Douglas R. Lloyd (U.S.A.), Professor E. A. Mason (U.S.A.), Dr. Stephen L. Matson (U.S.A.), Professor T. Matsuura (Canada), Dr. Scott B. McCray (U.S.A.), Professor Patrick Meares (United Kingdom), Dr. Ulrich Merten (U.S.A.), Dr. B. S. Minhas (U.S.A.), Professor Masayuki Nakagaki (Japan), Professor Tsutomu Nakagawa (Japan), Professor Richard D. Noble (U.S.A.), Professor Hisashi Odani (Japan), Mr. David J. Paulson (U.S.A.), Dr. John Pellegrino (U.S.A.), Professor E. Petropoulos (Greece), Dr. Ravi Prasad (U.S.A.), Dr. Ravi Prasad (U.S.A.), Dr. Pushpinder Puri (U.S.A.), Dr. Daryl L. Roberts (U.S.A.), Professor Charles E. Rogers (U.S.A.), Dr. Ian C. Roman (U.S.A.), Dr. E. S. Sanders (U.S.A.), Dr. R. D. Sanderson (Israel), Dr. William J. Schell (U.S.A.), Mr. John L. Short (U.S.A.), Professor Kamalesh K. Sirkar (U.S.A.), Professor Vivian Stannett (U.S.A.), Professor E. Staude (Germany), Professor S. Alexander Stern (U.S.A.), Professor Akihiko Tanioka (Japan), Dr. G. B. Tanny (Israel), Professor Gun M. Tragardh (Sweden), H. S. Mike Tseng (U.S.A.), Dr. Rich Ubersax (U.S.A.), Professor Tadashi Uragami (Japan), Dr. Venkat Venkataraman (U.S.A.), Professor Vladimir V. Volkov (Russia), Professor Y. Yampol'skii (Russia), Professor Meng Guang Yao (Belarus), Professor Masakazu Yoshikawa (Japan), Dr. Leos Zeman (U.S.A.)

CONTENTS

General Terms

1.   **asymmetric membrane:**   membrane constituted of two or more structural planes of non-identical morphologies (§18)

2.   **co-current flow:**   flow pattern through a membrane module in which the fluids on the upstream and the downstream sides of the membrane move parallel to the membrane surface and in the *same* directions (*Note:* see Fig. 1a)

3.   **completely-mixed (perfectly-mixed) flow:**   flow through a membrane module in which fluids on both the upstream and downstream sides of the membrane are individually well-mixed (*Note:* see Fig. 1b)

4.   **composite membrane:**   membrane having chemically or structurally distinct layers

5.   **continuous membrane column:**   membrane module(s) arranged in a manner to allow operation analogous to that of a distillation column, with each module acting as a stage

6.   **counter-current flow:**   flow through a membrane module in which the fluids on the upstream and the downstream sides of the membrane move parallel to the membrane surface but in *opposite* directions (*Note:* see Fig. 1c)

| (a) co-current flow | (b) completely-mixed flow | (c) counter-current flow |

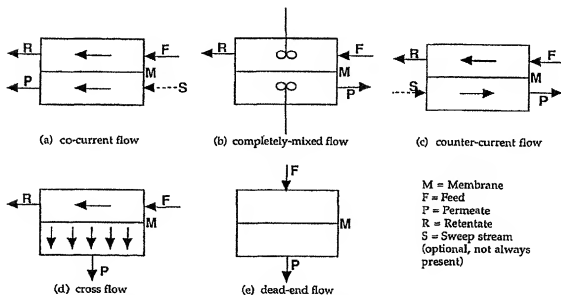| (d) cross flow | (e) dead-end flow | M = Membrane
F = Feed
P = Permeate
R = Retentate
S = Sweep stream
(optional, not always
present) |

Figure 1: Types of ideal continuous flows used in membrane-based separations

7.   **cross flow:** flow through a membrane module in which the fluid on the ypstream side of the membrane moves parallel to the membrane surface and the fluid on the downstream side of the membrane moves away from the membrane in the direction normal to the membrane surface (*Note:* see Fig. 1d)

8.   **dead-end flow:** flow through a membrane module in which the only outlet for upstream fluid is through the membrane (*Note:* see Fig. 1e)

9.   **dense (non-porous) membrane:** membrane with no detectable pores

10.   **downstream:**   side of a membrane from which permeate emerges

11.   **dry-phase separation membrane formation:** process in which a dissolved polymer is precipitated by evaporation of a sufficient amount of solvent to form a membrane structure (*Note:* Appropriate mixtures of additives are present in solution with the polymer to alter its precipitation tendency during solvent evaporation)

12.   **dry-wet phase separation membrane formation:** combination of the dry- (§11) and the wet-phase formation processes (§45)

13.   **dynamic membrane formation:** process in which an active layer is formed on the membrane surface by the deposition of substances contained in the fluid being treated

14.   **flux,** J_i, **[kmol m^{-2} s^{-1}]:** number of moles, volume, or mass of a specified component i passing per unit time through a unit of membrane surface area normal to the thickness direction. {*Note:* other commonly used units for J_i include [m^3 / m^{-2} s^{-1}], or [kg/m^{-2} s^{-1}] or [m^3 (measured at standard temperature and pressure) m^{-2} s^{-1}]}

15.   **fouling:** process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores

16.   **homogeneous membrane:** membrane with essentially the same structural and transport properties throughout its thickness

17.   **Langmuir-Blodgett (LB) membrane:** synthetic composite membrane formed by sequential depositing of one or more monolayers of surface-active component onto a porous or nonporous support

18.   **membrane:** structure, having lateral dimensions much greater than its thickness, through which mass transfer may occur under a variety of driving forces

19.   **membrane compaction:** compression of membrane structure due to a pressure difference across its thickness

20.   **membrane conditioning (pretreatment):** process carried out on a membrane after the completion of its preparation and prior to its use in a separation application (*Note 1:* thermal annealing to relieve stresses or pre-equilibration in a solution similar to the feed stream it will contact are examples of conditioning treatments) [*Note 2:* conditioning treatments differ from post-treatments (§25), since the latter occur before exposure to feed type solutions, while conditioning may occur using actual feed solutions]

21.   membrane distillation: distillation process in which the liquid and gas phases are separated by a porous membrane, the pores of which are not wetted by the liquid phase

22.   membrane module (cell): manifold assembly containing a membrane or membranes to separate the streams of feed, permeate, and retentate (*Note:* see Fig. 2a-c)

23.   membrane partition (distribution) coefficient: parameter equal to the equilibrium concentration of a component ($c_i^{(m)}$) in a membrane divided by the corresponding equilibrium concentration of the component in the external phase in contact with the membrane surface, $c_i^{(e)}$. $\left(\text{viz.,}\ K = c_i^{(m)} \middle/ c_i^{(e)} \right)$
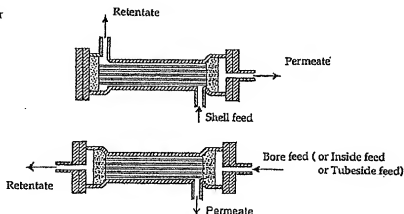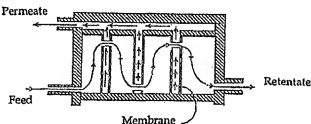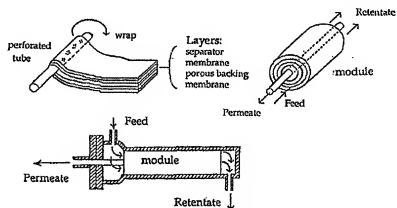

Figure 2: Types of modules used in membrane-based separation.

24.   **membrane physical aging:** change in the transport properties of a membrane over a period of time due to physical chemical structural alterations

25.   **membrane post-treatment:** process carried out on a membrane after its essential structure has been formed but *prior* to its exposure to an actual feed stream (§20.)

26.   **membrane reactor:** device for simultaneously carrying out a reaction and membrane-based separation in the same physical enclosure

27.   **penetrant (permeant):** entity from a phase in contact with one of the membrane surfaces that passes through the membrane

28.   **permeability coefficient,** P_i, [kmol m m^{-2} s^{-1} kPa^{-1}]: parameter defined as a transport flux, J_i, per unit transmembrane driving force per unit membrane thickness, viz., $\bar{P}_i = J_i$ / [(transmembrane driving force of component i/ℓ] {*Note:* other commonly used units for P_i include [m^3 m m^{-2} s^{-1} kPa^{-1}], [m^3 (measured at standard temperature and pressure m m^{-2} s^{-1} kPa^{-1}] or [kg m m^{-2} s^{-1} kPa^{-1}])

29.   **permeance (pressure normalized flux),** [kmol m^{-2} s^{-1} kPa^{-1}]: transport flux per unit transmembrane driving force, viz., P_1/ℓ (§28) {*Note:* other commonly used units include [m^3 m^{-2} s^{-1} kPa], [kg m^{-2} s^{-1} kPa^{-1}], or [m^3 (measured at standard temperature and pressure) m^{-2} s^{-1} kPa^{-1}]}

30.   **permeate:** stream containing penetrants that leaves a membrane module (*Note:* see Figs. 1 and 2)

31.   **perstraction:** separation process in which membrane permeation and extraction phenomena occur by contacting the downstream with an extracting solvent

32.   **relative recovery,** $\eta_{n,B}$ (substance efficiency): amount-of-substance of a component B collected in a useful product, $\eta_{B,out}$, divided by the amount-of-substance of that component entering the process, $\eta_{B,in}$: $\eta_{n,B} = \eta_{B,out}/\eta_{B,in}$ [*Note:* in membrane separations, the useful product may be either the retained material (or retentate) or the permeated material (or permeate)]

33.   **rejection factor, R:** parameter equal to one minus the ratio the concentrations of a component (i) on the downstream and upstream sides of a membrane [*Note 1:* R = 1 - [(c_i)downstream /(c_i)upstream)] [*Note 2:* concentrations may be either in the bulk ("Apparent Rejection Factor") or at the membrane surface ("Intrinsic Rejection Factor")] [*Note 3:* rejection factor refers to a local relationship between upstream and downstream concentrations while retention factor (§35) and relative recovery (§32) refers to feed and retentate or permeate leaving the module]

34.   **retentate (raffinate):** stream that has been depleted of penetrants that leaves the membrane modules without passing through the membrane to the downstream

35.   **retention factor,** r_F: parameter defined as one minus the ratio of permeate concentration to the retentate (§34) concentration of a component (*i*) {*Note 1:* $r_F = 1 - \left[(c_i)_p / (c_i)_k \right]$} [*Note 2.* "p" and "r" refer to permeate (§30) and retentate (§34). See Fig. 1] [*Note 3:* Compare rejection factor. (§33)]

36.   **selective membrane skin:** region, often located at the upstream face of an asymmetric membrane, that forms a thin, distinguishable layer primarily responsible for determining the permeability of the asymmetric membrane

37.   separation coefficient, $S_C(AB)$: ratio of the compositions of component A and B in the downstream relative to the ratio of compositions of these components in the upstream {*Note 1*: For example, if compositions are expressed in mole fractions (X_A and X_B), $S_C(AB) = [X_A/X_B]_{downstream} / [X_A/X_B]_{upstream}$} (*Note 2*: The separation coefficient can also be defined equivalently in terms of concentrations in the downstream and upstream, since only ratios are involved) [*Note 3*: The separation coefficient refers to a local relationship between concentrations on the upstream (§44) and downstream (§10) concentrations while the separation factor (§38) refers to retentate (§34) and permeate (§30) leaving the module]

38.   separation factor, $S_F(AB)$: ratio of the compositions of components A and B in the permeate relative to the composition ratio of these components in the retentate {*Note 1*: For example, $S_F(AB) = [X_A/X_B]_{Permeate} / [X_A/X_B]_{Retentate}$} (*Note 2*: The separation factor can also be defined in terms of concentrations in the permeate and retentate since only ratios are involved. *Note*: see Fig. 1) [*Note 3*. Compare separation coefficient. (§37)]

39.   sol-gel membrane formation: multistep process for making membranes by a reaction between two chemically multifunctional materials, dissolved in a solvent, that results in a network structure with solvent retained in the network followed by heat treatment to achieve a desired pore structure

40.   stage cut: parameter defined as the fractional amount of the total feed entering a membrane module that passes through the membrane as permeate (*Note*: see Fig. 1)

41.   synthetic (artificial) membrane: membrane formed by a process not occurring in nature

42.   thermally-induced phase-separation membrane formation: process in which a dissolved polymer is precipitated or coagulated by controlled cooling to form a membrane structure

43.   track-etch membrane formation: process for forming porous membranes with well-defined pores by exposing a dense film to ion bombardment followed by etching of the damaged region (*Note*: Usually produces pores with a narrow size distribution)

44.   upstream: side of a membrane into which penetrants enter from the feed stream

45.   wet-phase separation membrane formation: process in which a dissolved polymer is precipitated by immersion in a non-solvent bath to form a membrane structure

Carrier-Mediated (Facilitated) Separations

46.   anchored (bound) carrier: distinct species bonded chemically to fixed sites within a membrane for the purpose of increasing the selective sorption and flux of a specific component in a feed stream relative to all other components

47.   carrier complexation coefficient, K_c, [kmol m^{-3}]: parameter defined as the ratio of the rate constants for the second order complexation and first order decomplexation reaction between a carrier and a penetrant: viz., A + M = AM [*Note 1*: $K_c = k_c/k_d$, where A & M, resp., are a penetrant and a carrier site within a membrane (see note 2)] [*Note 2*: both anchored carrier sites (§46) and mobile carrier sites (§59) are possible]

48.   carrier complexation: phenomenon in which carrier molecules form a coordinated structure with penetrant molecules

49.   carrier deactivation: chemical transformations involving a carrier entity which render it less capable of undergoing the desired interaction with a penetrant

50.   carrier leaching: loss of carrier due to its partitioning by mass transport into one or both external phases

51.   carrier-mediated (facilitated) transport: process in which chemically distinct carrier species (§46, §59) form complexes with a specific component in the feedstream, thereby increasing the flux (§14) of this component relative to other components

52.   complexation rate constant, k_c [kmol^{-1} m^3 s^{-1}]: carrier complexation rate divided by the product of the local concentrations of the carrier and the complexable component, viz., k_c = (complexation rate)/[$(c)_{carrier}(c)_{complexable\ component}$] where concentrations are given in [kmol m^{-3}] and complexation rate is given in [kmol m^{-3} s^{-1}]

53.   coupled transport: process in which the flux of one component between the upstream and downstream is linked to the flux of a second component

54.   Damkohler number: dimensionless number equal to the characteristic time (ℓ^2/D_{AM}) for diffusion of complexed component across a membrane of thickness, ℓ, divided by the characteristic time (k_d^{-1}) for the decomplexation reaction between a carrier (M) and a complexed penetrant, A, viz., $\ell^2/(D_{AM}k_d)$ when D_{AM} is the effective diffusion coefficient of the complexed carrier entity in the membrane

55.   decomplexation rate constant, k_d [s^{-1}]: ratio of the decomplexation rate to the product of the local concentration of the complexed carrier, viz., k_d = (decomplexation rate) / $(c)_{complexed\ carrier}$ {Note: typical units for decomplexation rate are [kmol m^{-3} s^{-1}], and for complexed carrier are [kmol m^{-3}]}

56.   enhancement factor, $\mathcal{E}$: ratio of the flux of a component, i (§14) across a carrier-containing membrane divided by the transmembrane flux of the same component across an otherwise identical membrane without carrier

{Note: $\mathcal{E} = \left[(j_i)_{with\ carrier} / (j_i)_{without\ carrier} \right]$}

57.   facilitation factor: parameter equal to the enhancement factor (§56) minus one (Note: $\mathcal{F} = \mathcal{E} - 1$)

58.   liquid membrane: liquid phase existing either in supported or unsupported form that serves as a membrane barrier between two phases

59.   mobile carrier: distinct species moving freely within a membrane for the purpose of increasing the selective sorption and flux of a specific component in a feed stream relative to all other components

60.   uphill transport: process in which diffusion of a component occurs from a less concentrated feed stream to a more concentrated permeate stream

Dialysis, Nanofiltration, Ultrafiltration, and Microfiltration Separations

61.   **backflush:** temporary reversal of the direction of the permeate flow

62.   **bubble point:** pressure at which bubbles first appear on one surface of an immersed porous membrane as gas pressure is applied to the other surface

63.   **cake layer:** layer comprised of rejected particulate materials residing on the upstream face of a membrane

64.   **concentration polarization:** concentration profile that has a higher level of solute nearest to the upstream membrane surface compared with the more-or-less well-mixed bulk fluid far from the membrane surface

65.   **concentration factor:** ratio of the concentration of a component i in the retentate to the concentration of the same component in the feed {*Note 1:* $c_F = \left[(c_i)_{retentate} / (c_i)_{feed} \right]$} (*Note:* see Fig. 1)} [*Note 2:* Compare retention factor (§35)]

66.   **dialysis:** membrane process in which transport is driven primarily by concentration differences, rather than by pressure or electrical-potential differences, across the thickness of a membrane

67.   **dialysis permeability coefficient:** permeability coefficient (§28) based on a transmembrane driving force expressed in terms of the concentration difference of a given component

68.   **gel fouling layer:** highly swollen fouling layer comprising a three-dimensional, possibly network, structure residing at the surface of a membrane

69.   **hemodialysis:** dialysis process (§66) in which undesired metabolites and toxic by-products, such as urea and creatine, are removed from blood

70.   **hemofiltration:** ultrafiltration process (§76) in which undesired metabolites and toxic by-products, such as urea and creatine, are removed from blood

71.   **hindered transport:** combined partition, diffusion and convection process in which the effective partition, diffusion and viscous drag coefficients in a restricted environment depend upon the ratio of the effective radius of the penetrant molecule to that of the pore

72.   **microfiltration:** pressure-driven membrane-based separation process in which particles and dissolved macromolecules larger than 0.1 μm are rejected

73.   **molecular-weight cutoff:** molecular weight of a solute corresponding to a 90% rejection coefficient (§33) for a given membrane

74.   · **nanofiltration:** pressure-driven membrane-based separation process in which particles and dissolved molecules smaller than about 2 nm are rejected

75.   **ultrafiltration:** pressure-driven membrane-based separation process in which particles and dissolved macromolecules smaller than 0.1 μm and larger than about 2 nm are rejected

Electrically Mediated Separations

76.   anion-exchange membrane: membrane containing fixed cationic charges and mobile anions that can be exchanged with other anions present in an external fluid in contact with the membrane

77.   bipolar membrane: synthetic membrane containing two oppositely charged ion-exchanging layers in contact with each other

78.   cation-exchange membrane: membrane containing fixed anionic charges and mobile cations which can be exchanged with other cations present in an external fluid in contact with the membrane

79.   charge-mosaic membranes: synthetic membrane composed of two-dimensional or three-dimensional alternating cation- and anion-exchange channels throughout the membrane

80.   Donnan exclusion: reduction in concentration of *mobile* ions within an ion exchange membrane due to the presence of *fixed* ions of the same sign as the mobile ions

81.   electro-dialysis: membrane-based separation process in which ions are driven through an ion-selective membrane under the influence of an electric field

82.   electro-osmosis: process by which water is transported across the thickness of an anion-exchange (§76) or cation-exchange membrane (§78) under an applied electric field

83.   limiting current density: current density at which dramatic increases in resistance are observed in an ion exchange membrane system under the influence of an applied electric field between the upstream and downstream

Gas, Vapor and Pervaporation Separations

84.   ideal separation factor: parameter defined as the ratio of the permeability coefficient of component A to that of component B and equal to the "separation factor" (§37) where a perfect vacuum exists at the downstream membrane face for gas and vapor permeation systems

85.   pervaporation: membrane-based process in which the feed and retentate streams are both liquid phases while permeant emerges at the downstream face of the membrane as a vapor

86.   .solution-diffusion (sorption-diffusion): molecular-scale process in which penetrant is sorbed into the upstream membrane face from the external phase, moves by molecular diffusion in the membrane to the downstream face and leaves into the external gas, vapor or liquid phase in contact with the membrane

87.   sweep: nonpermeating stream directed past the downstream membrane face to reduce downstream permeant concentration

Reverse Osmosis Separations

88.   **brackish water:** term used to indicate water having a total dissolved-solids content that is less than that of sea water but above that of potable water

89.   **feed pretreatment:** process carried out on a crude feed stream, prior to feeding to a membrane separation system, to eliminate objectionable components such as biological agents and colloids that might impede the stable operation of the membrane

90.   **permeate post-treatment:** one or more final conditioning steps to improve permeate quality, e.g., contacting with anion exchange resins to remove trace ions in the permeate of a reverse osmosis product stream

91.   **potable water:** term used to indicate water having a total dissolved solids content of less than 500 ppm with a sufficiently low level of biological agents, suspended solids, organic odour- and colour-generating components to be safe and palatable for drinking

92.   **reverse osmosis:** liquid-phase pressure-driven separation process in which applied transmembrane pressure causes selective movement of solvent against its osmotic pressure difference

References

Audinos, R. and P. Isoard, eds., *Glossaire des termes techniques des procedes a membranes*, France: Societe Francaise de Filtration, 1986.

Glossary of Atmospheric Chemistry Terms, compiled by Jack G. Calvert, Applied Chemistry Division, Commission on Atmospheric Chemistry, IUPAC, 1990.

Porter, Mark, *Handbook of Industrial Membrane Technology*, Park Ridge, NJ: Noyes Publications, 1990.

Quantities, Units and Symbols in Physical Chemistry, I. M. Mills, et al., Blackwell Scientific, 1993.

Standard D1129-90, ASTM Committee on Water, Subcommittee on Membrane and Ion Exchange, D19.08, Vol. 11.01, April 1991.

Standard D5090, ASTM Committee on Water, Subcommittee on Membrane and Ion Exchange D19.080, Vol. 11.02, May 1991.

Terminology for Electrodialysis, prepared by Karl Hattenback, European Society of Membrane Science and Technology, issued November 1988.

Terminology for Membrane Distillation, prepared by A. C. M. Franken and S. Ripperger, University of Twente.

Terminology for Pressure Driven Membrane Operations, prepared by Vassilis Gekas, European Society of Membrane Science and Technology, issued June 1986.

Terminology in Pervaporation, prepared by K. W. Boddeker, European Society of Membrane Science and Technology, issued November 1989.

Microfiltration and Ultrafiltration

Principles and Applications

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sured surface porosities were more than an order of magnitude less than the bulk values for $\epsilon_b < 0.55$.

D. Gradient Anisotropy

Interesting MF/UF membrane structures can also be prepared with cross-sectional pore (void) size gradients. Their morphology displays a character of gradient anisotropy (nonuniformity or asymmetry). Figure 26a shows the cross-sectional SEM view (at $1520\times$ magnification) of a highly aniso-tropic PES UF membrane developed in the author's laboratory. Figure 26, b and c, shows the corresponding images of the upper (bath) surface (at $64,500\times$) and of the lower (belt) surface (at $4860\times$). The pore diameters in the upper surface are roughly in the range of $0.01–0.03$ µm (i.e., in the size range of macromolecules), while the large quasicircular pores in the
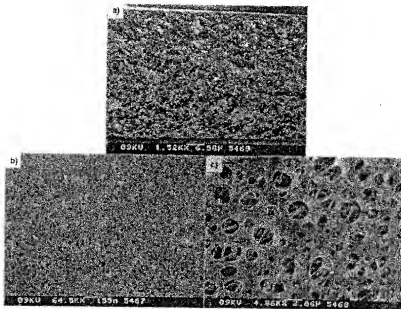
Figure 26 The gradient anisotropy character of the UF membrane morphol-ogy. SEM images of a polyethersulfone UF membrane with filtration cutoff of about 100,000 g/mol: (a) cross-section at $1520\times$, (b) top surface at $64,500\times$, and (c) bottom surface at $4860\times$. Degree of anisotropy, $A \cong 100$.

SYNTHETIC POLYMERIC MEMBRANES

A Structural Perspective

Second Edition

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pore former, albeit one of higher than usual MW. As such it *assisted* the membrane polymer to assume the Sol 2 micellar structure prior to gelation. Physical, *but not single* T_g, compatibility is required for practical PAPI blends. Too compatible a blend presumably leads to Sol 1 structures which gel as such without assuming the necessary Sol 2 configuration. PAPI membranes are usually skinless, isotropic with a narrow pore-size distribution, of intermediate porosity (~50%), and characterized by good to excellent mechanical properties. The choice of both membrane and leaching polymers for the PAPI process is governed by, the rules which are applicable to polymer blends which, unfortunately, given our present level of understanding, means that it is made on a largely empirical basis.

A potential application for PAPI process membranes is to serve as microporous supports for thin-film composites. This is particularly attractive in the *reverse-sequence* (RS) method for forming defect-free thin-film composites (see Section 7.7), where the thin film can be deposited prior to leaching the assisting polymer, thereby providing a dense impermeable surface which is ideal for coating. After a thin film of preformed-, or *in situ*-, formed polymers from fluid solutions has been deposited and/or cured, the support layer of the composite membrane can be made porous by leaching.

7.6 INHOMOGENEITY IN DEPTH

Prior to 1960, only isotropic or slightly anisotropic phase-inversion membranes were known. Today there are two types of inhomogeneity in depth which are of importance: *skinning* and *anisotropy*.

Skinning or asymmetry refers to a structure in which a thin (0.1–0.25 μm in depth) dense skin layer is integrally bonded in series with a thick (~100 μm) porous substructure. The skin layer determines both the permeability and perm-selectivity of the bilayer, whereas the porous substructure functions primarily as a physical support for the skin. The first skinned membrane, the wet-process cellulose acetate type developed by Loeb and Sourirajan[20] in 1960 for desalination by hyperfiltration, is universally acknowledged as the instrument which heralded the advent of the golden age of membranology. In the Loeb and Sourirajan, or *integrally-skinned*, membrane, skin and substructure are composed of the same material. Differences in density between the two layers are the result of interfacial forces and the fact that solvent loss occurs more rapidly from the air–solution and solution–nonsolvent bath interfaces than from the solution interior. Certain aspects of the skin layer remain a hotly debated issue. The earliest EM studies failed to discover any specific details of the microstructure, from which it was deduced that the skin was in an amorphous glassy state, similar to that of a solvent-cast bulk film. As we shall see, this conclusion was only partly true. Schultz and Asunmaa[21] discovered the presence of (~200 Å in diameter) spherical micelles in the skins of cellulose acetate membranes which had been etched with argon ions. Similar micelles were subsequently found by Kesting[6] in the skins of dry-process mem-

Basic Principles of Membrane Technology

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continuing, not only for membrane processes yet to reach the stage of commercialisation, but also for already existing membrane processes.

I.4.   Definition of a membrane

Although it is difficult to give an exact definition of a membrane, a general definition could be: *a selective barrier between two phases, the term 'selective' being inherent to a membrane or a membrane process.* It should be noted that this is a macroscopic definition while separation should be considered at the microscopic level. The definition says nothing about membrane structure nor membrane function.

        A membrane can be thick or thin, its structure can be homogeneous or heterogeneous, transport can be active or passive, passive transport can be driven by a pressure, concentration or a temperature difference. In addition, membranes can be natural or synthetic, neutral or charged. To obtain a more informative understanding, membranes can be classified according to different view points. The first classification is by nature, i.e. biological or synthetic membranes. This is the clearest distinction possible. It is also an essential first distinction since the two types of membranes differ completely in structure and functionality. Although this book emphasises synthetic membranes, a section in chapter II is also devoted to biological membranes. The latter can be subdivided into living and non-living membranes, and although living membranes are essential for life on earth they are not included here because this would increase the scope of this book to too great an extent. On the other hand, non-living biological membranes (liposomes and vesicles from phospholipids) are increasingly important in actual separation processes, especially in medicine and biomedicine. Synthetic membranes can be subdivided into organic (polymeric or liquid) and inorganic (ceramic, metal) membranes. Both types will be discussed in more detail in chapter III.

        Another means of classifying membranes is by morphology or structure. This is also a very illustrative route because the membrane structure determines the separation mechanism and hence the application. If we confine ourselves to solid synthetic membranes, two types of membrane may be distinguished, i.e. symmetric or asymmetric membranes. The two classes can be subdivided further as shown schematically in figure I - 5. The thicknesses of symmetric membranes (porous or nonporous) range roughly from 10 to 200 μm, the resistance to mass transfer being determined by the total membrane thickness. A decrease in membrane thickness results in an increased permeation rate.

        A breakthrough to industrial applications was the development of asymmetric membranes [25]. These consist of a very dense toplayer or skin with a thickness of 0.1 to 0.5 μm supported by a porous sublayer with a thickness of about 50 to 150 μm. These membranes combine the high selectivity of a dense membrane with the high permeation rate of a very thin membrane. Figure I - 6 depicts the cross-section of an asymmetric membrane in which the structural asymmetry is clearly visible. The resistance to mass transfer is